

In the Claims

1. (currently amended) A method of catalyzing an oxidation reaction, which comprises contacting an oxidizable substrate selected from the group consisting of textiles, hard surfaces, waste-printed paper, and pulp used in paper making, with an oxidizing agent selected from the group consisting of a peroxide and a peroxide forming substance, in the presence of a catalytically effective amount of at least one metal complex compound of formula (1)



wherein

Me is manganese, titanium, iron, cobalt, nickel or copper,

X is a coordinating or bridging radical,

n and m are each independently of the other an integer having a value of from 1 to 8,

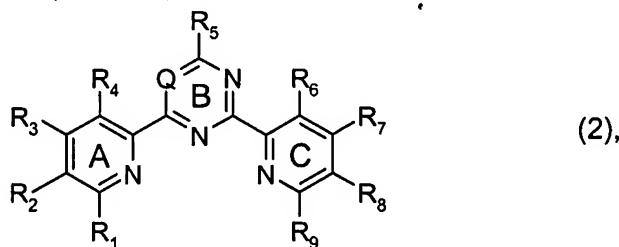
p is an integer having a value of from 0 to 32,

z is the charge of the metal complex,

Y is a counter-ion,

q = z/(charge of Y), and

L is a ligand of formula (2)



wherein

Q is N or CR₁₀,

R₁, R₂, R₃, R₄, R₅, R₆, R₇, R₈, R₉ and R₁₀ are each independently of the others hydrogen; unsubstituted or substituted C₁-C₁₈alkyl or unsubstituted or substituted aryl; cyano; halogen; nitro; -COOR₁₁ or -SO₃R₁₁ wherein

~~R₁₁ is in each case hydrogen, a cation or unsubstituted or substituted C₁-C₁₈alkyl or unsubstituted or substituted aryl; -SR₁₂, -SO₂R₁₂ or -OR₁₂ wherein~~

~~R₁₂ is in each case hydrogen or unsubstituted or substituted C₁-C₁₈alkyl or unsubstituted or substituted aryl; -NR₁₃R₁₄; -(C₁-C₆alkylene)-NR₁₃R₁₄; -N[⊕]R₁₃R₁₄R₁₅; -(C₁-C₆alkylene)-N[⊕]R₁₃R₁₄R₁₅; -N(R₁₂)-(C₁-C₆alkylene)-NR₁₃R₁₄; -N[(C₁-C₆alkylene)-NR₁₃R₁₄]₂; -N(R₁₂)-(C₁-C₆alkylene)-N[⊕]R₁₃R₁₄R₁₅; -N[(C₁-C₆alkylene)-N[⊕]R₁₃R₁₄R₁₅]₂; -N(R₁₂)-N-R₁₃R₁₄ or -N(R₁₂)-N[⊕]R₁₃R₁₄R₁₅, wherein~~

R₁₁ is in each case hydrogen, a cation or unsubstituted or substituted C₁-C₁₈alkyl or unsubstituted or substituted aryl.

R₁₂ is in each case hydrogen or unsubstituted or substituted C₁-C₁₈alkyl or unsubstituted or substituted aryl.

~~R₁₂ is as defined above and~~

~~R₁₃, R₁₄ and R₁₅ are each independently of the other(s) hydrogen or unsubstituted or substituted C₁-C₁₈alkyl or unsubstituted or substituted aryl, or~~

~~R₁₃ and R₁₄, together with the nitrogen atom linking them, form an unsubstituted or substituted 5-, 6- or 7-membered ring which may contain further hetero atoms.~~

2. (previously presented) A method according to claim 1, wherein Me is manganese in the oxidation state II, III, IV or V.

3. (canceled)

4. (previously presented) A method according to claim 1, wherein X is CH₃CN, H₂O, F⁻, Cl⁻, Br⁻, HOO[·], O₂²⁻, O²⁻, R₁₆COO[·], R₁₆O[·], LMeO[·] or LMeOO[·], wherein R₁₆ is hydrogen, -SO₃C₁-C₄alkyl or unsubstituted or substituted C₁-C₁₈alkyl or substituted or unsubstituted aryl, and L and Me are as defined in claim 1.

5. (previously presented) A method according to claim 1, wherein Y is R₁₇COO[·], ClO₄⁻, BF₄⁻, PF₆⁻, R₁₇SO₃⁻, R₁₇SO₄²⁻, SO₄²⁻, NO₃⁻, F⁻, Cl⁻, Br⁻, I⁻, citrate, tartrate or oxalate, wherein R₁₇ is hydrogen or unsubstituted or substituted C₁-C₁₈alkyl or substituted or unsubstituted aryl.

6. (previously presented) A method according to claim 1, wherein n is an integer having a value of from 1 to 4.

7. (previously presented) A method according to claim 1, wherein m is an integer having a value of 1 or 2.

8. (previously presented) A method according to claim 1, wherein p is an integer having a value of from 0 to 4.

9. (previously presented) A method according to claim 1, wherein z is an integer having a value of from 8- to 8+.

10. (previously presented) A method according to claim 1, wherein aryl is phenyl or naphthyl each unsubstituted or substituted by C₁-C₄alkyl, C₁-C₄alkoxy, halogen, cyano, nitro, carboxy, sulfo, hydroxy, amino, N-mono- or N,N-di-C₁-C₄alkylamino unsubstituted or substituted by hydroxy in the alkyl moiety, N-phenylamino, N-naphthylamino, phenyl, phenoxy or by naphthyoxy.

11. (previously presented) A method according to claim 1, wherein the 5-, 6- or 7-membered ring formed by R₁₃ and R₁₄ together with the nitrogen atom linking them is an unsubstituted or C₁-C₄alkyl-substituted pyrrolidine, piperidine, piperazine, morpholine or azepane ring wherein the nitrogen atoms may be quaternised.

12. (currently amended) A method according to claim 1, wherein R₅ is C₁-C₁₂alkyl; phenyl unsubstituted or substituted by C₁-C₄alkyl, C₁-C₄alkoxy, halogen, cyano, nitro, carboxy, sulfo, hydroxy, amino, N-mono- or N,N-di-C₁-C₄alkylamino unsubstituted or substituted by hydroxy in the alkyl moiety, N-phenylamino, N-naphthylamino, phenyl, phenoxy or by naphthyoxy; cyano; halogen; nitro; -COOR₁₁ or -SO₃R₁₁ wherein

~~R₄₄ is in each case hydrogen, a cation, C₁-C₄alkyl, unsubstituted phenyl or phenyl substituted as indicated above; -SR₁₂, -SO₂R₁₂ or -OR₁₂ wherein~~

~~R₄₂ is in each case hydrogen, C₁-C₄alkyl, unsubstituted phenyl or phenyl substituted as indicated above; -NR₁₃R₁₄; -(C₁-C₆alkylene)-NR₁₃R₁₄; -N[⊕]R₁₃R₁₄R₁₅; -(C₁-C₆alkylene)-N[⊕]R₁₃R₁₄R₁₅; -N(R₁₂)-(C₁-C₆alkylene)-NR₁₃R₁₄; -N(R₁₂)-(C₁-C₆alkylene)-N[⊕]R₁₃R₁₄R₁₅; -N(R₁₂)-N-R₁₃R₁₄ or -N(R₁₂)-N[⊕]R₁₃R₁₄R₁₅, wherein~~

R₁₁ is in each case hydrogen, a cation, C₁-C₁₂alkyl, unsubstituted phenyl or phenyl substituted as indicated above,

R₁₂ is in each case hydrogen, C₁-C₁₂alkyl, unsubstituted phenyl or phenyl substituted as indicated above,

~~R₄₂ may have one of the above meanings and~~

R₁₃, R₁₄ and R₁₅ are each independently of the other(s) hydrogen, unsubstituted or hydroxy-substituted C₁-C₁₂alkyl, unsubstituted phenyl or phenyl substituted as indicated above, or R₁₃ and R₁₄, together with the nitrogen atom linking them, form a pyrrolidine, piperidine, piperazine, morpholine or azepane ring unsubstituted or substituted by at least one unsubstituted C₁-C₄alkyl and/or substituted C₁-C₄alkyl, wherein the nitrogen atom may be quaternised, and R₁, R₂, R₃, R₄, R₆, R₇, R₈, R₉ and R₁₀ may be as defined in claim 1 or are hydrogen.

13. (currently amended) A method according to claim 1, wherein

R₅ is phenyl unsubstituted or substituted by C₁-C₄alkyl, C₁-C₄alkoxy, halogen, phenyl or by hydroxy; cyano; nitro; -COOR₁₁ or -SO₃R₁₁ wherein

~~R₄₄ is in each case hydrogen, a cation, C₁-C₄alkyl or phenyl; -SR₁₂, -SO₂R₁₂ or -OR₁₂ wherein~~

~~R₄₂ is in each case hydrogen, C₁-C₄alkyl or phenyl; -N(CH₃)-NH₂ or -NH-NH₂; amino; N-mono- or N,N-di-C₁-C₄alkylamino unsubstituted or substituted by hydroxy in the alkyl moiety; or an unsubstituted or C₁-C₄alkyl-substituted pyrrolidine, piperidine, piperazine, morpholine or azepane ring, wherein~~

R₁₁ is in each case hydrogen, a cation, C₁-C₄alkyl or phenyl and

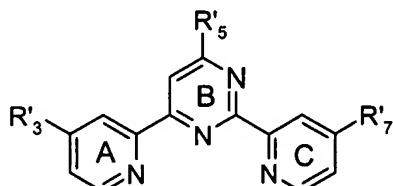
R₁₂ is in each case hydrogen, C₁-C₄alkyl or phenyl.

14. (previously presented) A method according to claim 1, wherein

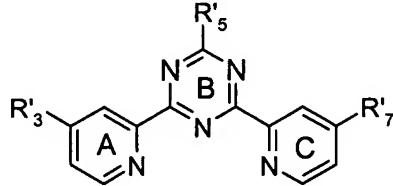
R_5 in L is C_1 - C_4 alkoxy; hydroxy; hydrazine; amino; N-mono- or N,N-di- C_1 - C_4 alkylamino unsubstituted or substituted by hydroxy in the alkyl moiety; or an unsubstituted or C_1 - C_4 alkyl-substituted pyrrolidine, piperidine, piperazine, morpholine or azepane ring.

15. (previously presented) A method according to claim 12, wherein R_1 , R_2 , R_3 , R_4 , R_6 , R_7 , R_8 , R_9 and R_{10} in L have the definitions given for R_5 in claim 12, but those radicals may additionally be hydrogen.

16. (previously presented) A method according to claim 1, wherein L is a compound of formula (3a) and/or (3b)



(3a)



(3b)

wherein R'_3 , R'_5 and R'_7 have the definitions given in claim 1 for R_3 , R_5 and R_7 .

17. (currently amended) A method according to claim 1, which comprises the use, as a catalyst for said oxidation reactions, of at least one metal complex compound of formula (1')



wherein

Me is manganese, titanium, iron, cobalt, nickel or copper,

X is a coordinating or bridging radical,

n and m are each independently of the other an integer having a value of from 1 to 8,

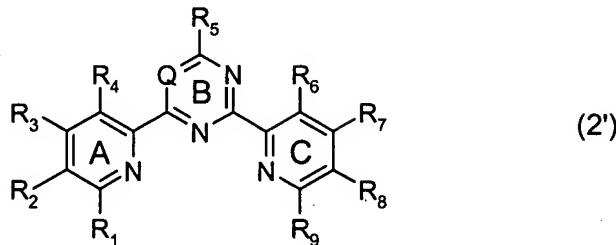
p is an integer having a value of from 0 to 32,

z is the charge of the metal complex,

Y is a counter-ion,

$q = z/(\text{charge of } Y)$, and

L' is a ligand of formula (2')



wherein

Q is N or CR_{10} ,

$R_1, R_2, R_3, R_4, R_5, R_6, R_7, R_8, R_9$ and R_{10} are each independently of the others hydrogen; unsubstituted or substituted C_1 - C_{18} alkyl or unsubstituted or substituted aryl; cyano; halogen; nitro; $-COOR_{11}$ or $-SO_3R_{11}$ wherein

R_{11} is in each case hydrogen, a cation or unsubstituted or substituted C_1 - C_{18} alkyl or substituted or unsubstituted aryl; $-SR_{12}$, $-SO_2R_{12}$ or $-OR_{12}$ wherein

R_{12} is in each case hydrogen or unsubstituted or substituted C_1 - C_{18} alkyl or unsubstituted or substituted aryl; $-NR_{13}R_{14}$; $-(C_1-C_6\text{alkylene})-NR_{13}R_{14}$; $-N^{\oplus}R_{13}R_{14}R_{15}$; $-(C_1-C_6\text{alkylene})-N^{\oplus}R_{13}R_{14}R_{15}$; $-N(R_{12})-(C_1-C_6\text{alkylene})-NR_{13}R_{14}$; $-N[(C_1-C_6\text{alkylene})-NR_{13}R_{14}]_2$; $-N(R_{12})-(C_1-C_6\text{alkylene})-N^{\oplus}R_{13}R_{14}R_{15}$; $-N[(C_1-C_6\text{alkylene})-N^{\oplus}R_{13}R_{14}R_{15}]_2$; $-N(R_{12})-N-R_{13}R_{14}$ or $-N(R_{12})-N^{\oplus}R_{13}R_{14}R_{15}$, wherein

R_{11} is in each case hydrogen, a cation or unsubstituted or substituted C_1 - C_{18} alkyl or substituted or unsubstituted aryl,

R_{12} is in each case hydrogen or unsubstituted or substituted C_1 - C_{18} alkyl or unsubstituted or substituted aryl,

R_{12} is as defined above and

R_{13} , R_{14} and R_{15} are each independently of the other(s) hydrogen or unsubstituted or substituted C_1 - C_{18} alkyl or unsubstituted or substituted aryl, or

R_{13} and R_{14} , together with the nitrogen atom linking them, form an unsubstituted or substituted 5-, 6- or 7-membered ring which may contain further hetero atoms,

with the proviso that

at least one of the substituents R_1 to R_{10} contains a quaternised nitrogen atom that is not bonded directly to one of the three rings A, B and/or C.

18. (previously presented) A method according to claim 17, wherein R_5 is not hydrogen.

19. (currently amended) A method according to claim 17, wherein

R_5 in L' is phenyl unsubstituted or substituted by C_1 - C_4 alkyl, C_1 - C_4 alkoxy, halogen, phenyl or by hydroxy; cyano; nitro; $-COOR_{11}$ or $-SO_3R_{11}$, wherein

R_{11} is in each case hydrogen, a cation, C_1-C_4 alkyl or phenyl; $-SR_{12}$, $-SO_2R_{12}$ or $-OR_{12}$ wherein R_{12} is in each case hydrogen, C_1-C_{14} alkyl or phenyl; $-N(CH_3)-NH_2$ or $-NH-NH_2$; amino; N-mono- or N,N-di- C_1-C_4 alkylamino unsubstituted or substituted by hydroxy in the alkyl moiety, wherein the nitrogen atoms may be quaternised; N-mono- or N,N-di- C_1-C_4 alkyl- $N^+R_{13}R_{14}R_{15}$ unsubstituted or substituted by hydroxy in the alkyl moiety, wherein

R_{11} is in each case hydrogen, a cation, C_1 - C_4 alkyl or phenyl.

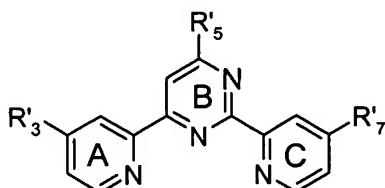
R_{12} is in each case hydrogen, C_1 - C_{14} alkyl or phenyl, and

R_{13} , R_{14} and R_{15} are each independently of the others hydrogen or unsubstituted or hydroxy-substituted C_1 - C_{12} alkyl, unsubstituted phenyl or phenyl substituted as indicated above, or

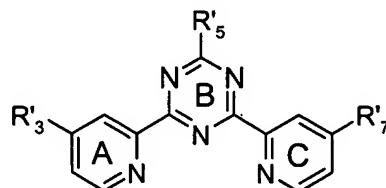
R_{13} and R_{14} , together with the nitrogen atom linking them, form a pyrrolidine, piperidine, piperazine, morpholine or azepane ring unsubstituted or substituted by at least one unsubstituted C_1 - C_4 alkyl and/or substituted C_1 - C_4 alkyl, wherein the nitrogen atom may be quaternised; N-mono- or N,N-di- C_1 - C_4 alkyl- $NR_{13}R_{14}$ unsubstituted or substituted by hydroxy in the alkyl moiety, wherein

R_{13} and R_{14} may be as defined above.

20. (previously presented) A method according to claim 17, wherein L' is a compound of formula (3'a) and/or (3'b)



(3'a)

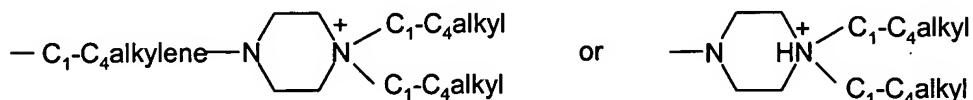


(3'b)

wherein R'₃, R'₅ and R'₇ have the definitions indicated in claim 17 for R₅, but R'₃ and R'₇ may additionally be hydrogen.

21. (previously presented) A method according to claim 20, wherein

(i) at least one of the substituents R'₃, R'₅ and R'₇ is one of the radicals



wherein the unbranched or branched alkylene group may be unsubstituted or substituted, and wherein the alkyl groups, which are unbranched or branched independently of one another, may be unsubstituted or substituted and wherein the piperazine ring may be unsubstituted or substituted.

22. (canceled)

23. (previously presented) A method according to claim 17, wherein L' contains precisely 1, 2 or 3 quaternised nitrogen atoms.

24-34. (canceled)